# Modifying the variational principle in the action integral functional derivation of time-dependent density functional theory

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## Abstract

According to a recent paper by G. Vignale [Phys. Rev. A **77**, 062511 (2008)], the problems arising in the original derivation of time-dependent density functional theory (TDDFT) based on the Runge-Gross (RG) action-integral functional (AIF) are due to an incorrect variational principle (VP). This argument and the proposed modification of the VP are critically analyzed. The more fundamental problem, though, is the indefiniteness of the RG AIF. In contrast to a widely held belief, that indefiniteness is not eliminated in the variational procedure, which unwittingly is corroborated by Vignale's initial point.

## I. INTRODUCTION

The original foundation of time-dependent density-functional theory (TDDFT) by Runge and Gross (RG) [1] was based on a variational principle for the so-called action integral functional (AIF). It was later found that this route to TDDFT leads to a violation of causality in the kernel of the exchange correlation functional ("causality paradox") [2–4] indicating a serious problem with the RG AIF. Van Leeuwen [5] located the problem in the arbitrary purely time-dependent (td) phase factor in the wave function that is not determined by the mapping between td densities and wave functions according to the first Runge-Gross theorem (RG1). As a consequence, the RG AIF is ill-defined (see also Ref. [6]).

While apparently van Leeuwen's analysis was not accepted unanimously in the TDDFT community, there was a broad consensus that any flaws in the original foundation would not affect the essence of TDDFT, because the td Kohn-Sham equations at the core of the theory could be established directly via the RG1 mapping theorem, evading the need for a variational principle. For a discussion of the problems arising in the latter mapping-based foundation the reader is referred to Ref. [6, 7].

In a recent paper [8], henceforth referred to as paper I, G. Vignale has attempted to rehabilitate the RG AIF and the TDDFT foundation associated with it. Guided by a correct finding, he concluded that the original formulation of the variational principle is wrong and must be modified appropriately. In the following brief report we will analyze Vignale's work in some detail. As will be seen, even when the problem of the definiteness of the RG AIF is ignored, the diagnosis in I is unfounded and the conclusions derived thereof not compelling.

## II. DEFINITION OF THE ACTION INTEGRAL FUNCTIONAL

Let us first address the problem of the indefiniteness of the RG AIF reading

$$A[n] = \int_{t_1}^{t_2} dt \, \langle \Psi[n](t) | i\frac{\partial}{\partial t} - \hat{H}(t) | \Psi[n](t) \rangle \tag{1}$$

Here $\Psi[n](t)$ is the wave function associated with the time-dependent (td) density $n(\boldsymbol{r}, t)$ according to the RG1 theorem. It should be noted that the RG1 mapping depends also on the initial value of the wave function, $\Psi(t_1)$; as usual, the initial state dependence will not explicitly be indicated for notational brevity.

As correctly stated by RG, the wave function $\Psi[n](t)$ is determined by the density only up to a purely time-dependent phase factor $e^{i\alpha(t)}$. However, the obvious consequence of this indefiniteness for the AIF has been falsely addressed in Ref. [1] and disregarded in later work (see, for example, Gross and Kohn [9]). Because of the time derivative in the integrand of the AIF, there arises a constant additive term, $c = -\int_{t_1}^{t_2} \dot{\alpha}(t)dt = \alpha(t_1) - \alpha(t_2)$. As the phase function $\alpha(t)$ is completely arbitrary so is the constant c. This means that the AIF, being a mapping of time-dependent densities (or density trajectories) onto mere numbers, is ill-defined.

Unfortunately, this finding is often dismissed by stating that the additive constant is "immaterial" with regard to the variational condition (see Casida [10]). A similar remark is in the brief Sec. IV of paper I: "While the absolute numerical value of the RG action has no physical meaning (because a multiplication of the wave function by an arbitrary phase factor changes its value by an arbitrary amount), it must be borne in mind that the action determines the dynamics through its variations, and those variations are independent of the arbitrary additive constant." It is noted (in parantheses) that "a similar situation occurs in classical mechanics, since the Lagrangian is defined up to an arbitrary total derivative with respect to time."

However, the latter remark is not quite accurate. The variational principle in classical mechanics is based on the action integral

$$S = \int_{t_1}^{t_2} dt\, L(q(t), \dot{q}(t), t) \tag{2}$$

assuming for simplicity only one degree-of-freedom. The requirement $\delta S = 0$ for variations of the coordinate function $\delta q(t)$ with $\delta q(t_1) = \delta q(t_2) = 0$ allows one to derive the well-known (Euler) equation-of-motion (EOM)

$$\frac{d}{dt}\frac{\partial L}{\partial \dot{q}} - \frac{\partial L}{\partial q} = 0 \tag{3}$$

The correct statement with regard to the non-uniqueness of the Lagrangian reads that $L$ is defined up to the total time derivative of an arbitrary function of the coordinates and time, say $\frac{d}{dt}f(q(t), t)$. The time integration leads to an additive constant $f(q(t_2), t_2) - f(q(t_1), t_1)$, for which the variation with respect to $\delta q(t)$ vanishes due to the requirement that $\delta q(t_1) = \delta q(t_2) = 0$, so that the result of the variational procedure is not affected. Obviously, the non-uniqueness associated with terms of the type $\frac{d}{dt}f(q(t), t)$ is quite specific and differs

from the vague formulation in paper I. As one can readily see, an arbitrary total derivative with respect to time leads to

$$\delta S = \int_{t_1}^{t_2} dt \, (\frac{\partial L}{\partial q} - \frac{d}{dt}\frac{\partial L}{\partial \dot{q}})\delta q(t) + C = 0 \tag{4}$$

Obviously, the arbitrary constant $C$ does not allow one to derive a meaningful EOM.

In the case of the RG AIF the argument mentioned above insinuates that $\delta c = 0$. As will be discussed at some detail below, the main point made in paper I is that the variation of the wave function, $\delta\Psi(t)$, associated with the variation of the underlying density, $n(t) + \delta n(t)$ (where $\delta n(t_1) = \delta n(t_2) = 0$), vanishes for the initial time $t_1$ but not for the final time $t_2$. This means, in particular, that the variation of the purely td phase will vanish at $t_1$ but not at $t_2$, that is, $\delta\alpha(t_2) \neq 0$. Obviously, the problem of the indefinite constant does not disappear in the variational procedure, and one cannot escape the conclusion that the RG AIF is ill-defined due to the ambiguity with respect to a purely time-dependent phase factor. It might be tempting to dismiss that innocuous phase factor in the wave function as somehow irrelevant, but one should recall that the time-dependence of stationary states consists exactly in such phase factors. They are an indispensable ingredient in the solutions of the td Schrödinger equation (SE).

Nothing meaningful can be deduced if the origin, that is, the RG AIF is ill-defined, and we could stop at this point. Nevertheless it may be of interest to elaborate on Vignale's attempt to modify the VP based on the AIF without dismissing the latter from the outset. So, tentatively, we will assume in the following that the RG AIF is a well-defined entity. Of course, the existence (definiteness) of the RG AIF is a concrete mathematical assertion being either true or false. A way to prove a premise false is to inspect its implications to be deduced in a logically stringent way. In this respect it is important to analyze arguments in paper I.

## III. VARIATIONAL PRINCIPLE FOR TIME-DEPENDENT QUANTUM MECHANICS

For a better understanding of the argumentation in paper I and clarifying certain misconceptions arising there, a brief review of the td variational principles (VP) in quantum mechanics will be given here. Of course, all of the following is well-known and a reader

familiar with the topic might safely skip this section.

The time-evolution of a wave function is determined by the td SE, together with an initial-state condition specifying the wave function at a given time. Often the exact solution of the td SE is not feasible, and one has to resort to approximations, say, to a parametrized form of the wave function, in which the parameters are time-dependent. The question is how to obtain a suitable EOM in such a situation. For this purpose one may resort to the well-known VPs of td quantum mechanics [11–14]. In the so-called Lagrangian form (see Ref. [13]), the VP is based on the action-type integral

$$A[\Psi] = \int_{t_1}^{t_2} dt \langle \Psi(t)| i\frac{\partial}{\partial t} - \hat{H}|\Psi(t)\rangle \tag{5}$$

for the time-development of a wave function in a given (though arbitrary) time interval $[t_1, t_2]$. The aim is to determine an "optimal" wave function $\Psi(t)$ (more accurately, derive a pertinent EOM) such that the action integral is stationary with respect to (small) variations of the form $\tilde{\Psi}(t) = \Psi(t) + \delta\Psi(t)$. Here the variations are required to vanish at the boundaries of the time interval, that is, $\delta\Psi(t_1) = \delta\Psi(t_2) = 0$. The VP then reads

$$\delta A[\Psi] = 0, \text{ where } \delta\Psi(t_1) = \delta\Psi(t_2) = 0 \tag{6}$$

The integrand in the action integral (5) can be interpreted as the expectation value of the deviation with respect to the exact td SE. The obvious idea underlying the VP (6) is that the desired wave function $\Psi(t)$ should minimize (make stationary) this deviation within the given time interval.

A more convenient, namely, instantaneous form of the td VP may be obtained by rewriting $\delta A[\Psi]$ in such a way that the VP applies directly to the integrand in the time-integration rather than to the action integral. Starting from

$$\delta A[\Psi] = \int_{t_1}^{t_2} dt\, \langle \delta\Psi(t)| i\frac{\partial}{\partial t} - \hat{H}|\Psi(t)\rangle + \int_{t_1}^{t_2} dt\, \langle \Psi(t)| i\frac{\partial}{\partial t} - \hat{H}|\delta\Psi(t)\rangle \tag{7}$$

one may replace the term involving $\delta\dot{\Psi}(t)$ using integration by parts,

$$\langle \Psi(t)|\delta\dot{\Psi}(t)\rangle = \frac{d}{dt}\langle \Psi(t)|\delta\Psi(t)\rangle - \langle \dot{\Psi}(t)|\delta\Psi(t)\rangle \tag{8}$$

Because $\delta\Psi(t_1) = \delta\Psi(t_2) = 0$ the integral of the total time derivative vanishes, and one arrives at

$$\delta A[\Psi] = 2\int_{t_1}^{t_2} dt\, Re\, \langle \delta\Psi(t)| i\frac{\partial}{\partial t} - \hat{H}|\Psi(t)\rangle = 0 \tag{9}$$

Taking into account that the integration boundaries $t_1, t_2$ are arbitrary, one may conclude (see Kucar *et al.* [15]) that the integral (9) vanishes if and only if

$$Re\,\langle\delta\Psi(t)|i\frac{\partial}{\partial t}-\hat{H}|\Psi(t)\rangle=0 \tag{10}$$

This establishes an instantaneous form of a td VP being fully equivalent to the Lagrangian form according to Eqs. (5,6). If together with the variations $\delta\Psi(t)$ also the variations $i\delta\Psi(t)$ are permitted, one arrives at the related form

$$Im\,\langle\delta\Psi(t)|i\frac{\partial}{\partial t}-\hat{H}|\Psi(t)\rangle=0 \tag{11}$$

Both forms are comprised in the well-known Dirac-Frenkel VP [11, 14], reading

$$\langle\delta\Psi|i\frac{\partial}{\partial t}-\hat{H}|\Psi\rangle=0 \tag{12}$$

This shows that the Lagrangian and the Dirac-Frenkel variational principles are equivalent provided that the both $\delta\Psi(t)$ and $i\delta\Psi(t)$ are allowed variations of the wave function. For an illustrative application of the Dirac-Frenkel VP the reader is referred to the derivation of the multi-configuration time-dependent Hartree (MCTDH) equations by Beck *et al.* [16].

Obviously, an exact solution of the td SE fulfills the Dirac-Frenkel (and the Lagrangian) VP. The converse proposition, however, is not true: In general, a wave function fulfilling the td VP need not be the exact solution of the td SE. As mentioned above, the purpose of the td VPs is to generate an EOM for cases where one cannot apply the td SE.

## IV. DENSITY-BASED TIME-DEPENDENT VARIATIONAL PRINCIPLE

Supposing that the RG AIF (1) is well-defined, we will now briefly discuss how, at least in principle, a valid EOM for an optimal (or exact) td density would emerge from the pertinent VP,

$$\delta A[n]=\int_{t_1}^{t_2}dt\,\langle\delta\Psi[n,\delta n](t)|i\frac{\partial}{\partial t}-\hat{H}|\Psi[n](t)\rangle+\int_{t_1}^{t_2}dt\,\langle\Psi[n](t)|i\frac{\partial}{\partial t}-\hat{H}|\delta\Psi[n,\delta n](t)\rangle=0 \tag{13}$$

Here the wave function $\Psi[n](t)$ is assumed to be somehow parametrized in terms of an underlying td density function $n(t)$. Once again, the intuitive idea of the VP is to minimize (make stationary) the expectation value of the td SE difference operator in a given time interval with respect to the manifold of permitted td densities (density trajectories). The

optimal td density $n(t)$ is to be compared to deviating density trajectories $\tilde{n}(t) = n(t)+\delta n(t)$, where $\delta n(t_1) = \delta n(t_2) = 0$. The variations of the td density induces associated variations of the wave function, reading (by definition)

$$\delta\Psi[n, \delta n](t) \equiv \Psi[n + \delta n](t) - \Psi[n](t) \tag{14}$$

Note that $\delta\Psi[n, \delta n](t)$ will depend both on $n(t)$ and $\delta n(t)$. While $\delta\Psi[n, \delta n](t)$ vanishes at the initial time $t_1$, this is not necessarily so at the final time, $t_2$, as Vignale rightly observes. We will come back to this point below.

Now the question is how to proceed from here to an at least formally compelling EOM for the optimal td density. For this purpose one might consider the possibility that $\delta A[n]$ can be written in the form of a td functional derivative with respect to the density at time $t$,

$$\delta A[n] = \int d\boldsymbol{r} \int_{t_1}^{t_2} dt f[n](\boldsymbol{r}, t)\delta n(\boldsymbol{r}, t) = 0 \tag{15}$$

where

$$f[n](\boldsymbol{r}, t) = \frac{\delta A[n]}{\delta n(\boldsymbol{r}, t)} \tag{16}$$

This would lead directly to the simple equation [1]

$$\frac{\delta A[n]}{\delta n(\boldsymbol{r}, t)} = 0 \tag{17}$$

However, such an outcome certainly is too simplistic. Even if one assumes that the wave function at a given time $t$ is determined by the density $n(t)$ at that time, the first time derivative $\dot{\Psi}(t)$ and thus also $\dot{n}(t)$ is needed to evaluate the expectation value of the td SE difference operator at time $t$. Therefore one has to expect that also a functional derivative with respect to $\dot{n}(t)$ will come into play leading to an expression of the type

$$\delta A[n] = \int d\boldsymbol{r} \int_{t_1}^{t_2} dt(f[n, \dot{n}](\boldsymbol{r}, t)\delta n(\boldsymbol{r}, t) + g[n, \dot{n}](\boldsymbol{r}, t)\delta\dot{n}(\boldsymbol{r}, t)) = 0 \tag{18}$$

where $f[n, \dot{n}](\boldsymbol{r}, t) = \frac{\delta A[n]}{\delta n(\boldsymbol{r},t)}$ and $g[n, \dot{n}](\boldsymbol{r}, t) = \frac{\delta A[n]}{\delta \dot{n}(\boldsymbol{r},t)}$ are functional derivatives with respect to $n(t)$ and $\dot{n}(t)$, respectively. Using integration by parts (as in the derivation of the classical Euler equations) and $\delta n(t_1) = \delta n(t_2) = 0$ it follows that

$$\delta A[n] = \int d\boldsymbol{r} \int_{t_1}^{t_2} dt \left( f[n, \dot{n}](\boldsymbol{r}, t) - \frac{d}{dt} g[n, \dot{n}](\boldsymbol{r}, t) \right) \delta n(\boldsymbol{r}, t) = 0 \tag{19}$$

yielding the Euler-type equation

$$f[n,\dot{n}](\boldsymbol{r},t) - \frac{d}{dt}g[n,\dot{n}](\boldsymbol{r},t) = 0 \tag{20}$$

In fact, the latter equation might be viewed as a viable EOM. Its practical use, however, would require a guess for an AIF depending not only on $n$ but also on $\dot{n}$.

Still, this may not yet be the end of the line. As was discussed in Ref. [6], one may need $\dot{n}(t)$ to determine $\Psi(t)$, so that the evaluation of the AIF integrand at a given time $t$ would even require the second time derivative $\ddot{n}(t)$. The corresponding expansion of $\delta A[n]$ would read

$$\delta A[n] = \int d\boldsymbol{r} \int_{t_1}^{t_2} dt \left[ \left( f[n,\dot{n},\ddot{n}](\boldsymbol{r},t) - \frac{d}{dt}g[n,\dot{n},\ddot{n}](\boldsymbol{r},t) \right) \delta n(\boldsymbol{r},t) + h[n,\dot{n},\ddot{n}](\boldsymbol{r},t)\delta\ddot{n}(\boldsymbol{r},t) \right] = 0 \tag{21}$$

where $h[n,\dot{n},\ddot{n}](\boldsymbol{r},t) = \frac{\delta A[n]}{\delta\ddot{n}(\boldsymbol{r},t)}$. In the latter case it is not clear how a viable EOM could be deduced at all.

To conclude, even if the RG AIF were well-defined it would not be guaranteed that a valid EOM for the td density can be deduced. Wether this is possible or not, depends crucially on a further specification of the RG mapping $n(t) \to \Psi[n](t)$. What pieces of information are needed to generate the wave function at a given time $t$ from the density trajectory? Does $n(t)$ suffice, are both $n(t)$ and $\dot{n}(t)$ needed, or is the situation even more complex, requiring, e.g., the past or even the future of the density trajectory with regard to $t$? So far, these questions have never been properly addressed let alone clarified.

## V. MODIFICATION OF THE VARIATIONAL PRINCIPLE?

After the preceding considerations we are now prepared to review the proposal for a modification of the AIF VP as presented in the basic Sec. II of paper I. Equations in I will be indicated here by adding the prefix I- to the original number.

In the beginning the td VP in the Lagrangian form (5) is considered, implying that this VP is equivalent to the td SE. As we have argued above the latter statement is misleading. Of course, the VP is based on the td SE in that the expectation value of the td SE difference operator is to be minimized (in a given time interval). As a result, a solution of the td SE fulfills the td VP, but the converse is not true: a wave function fulfilling the td VP must not necessarily be a solution of the td SE. The "proof" given in I (second column of p. 2) for

that incorrect assertion proceeds in the familiar way used to demonstrate the equivalence of the Lagrangian and Dirac-Frenkel VPs (see above), but ends after the time integration by parts with the following equation (I-6):

$$\delta A[\Psi] = \int_{t_1}^{t_2} dt\, \langle \delta\Psi(t)|i\frac{\partial}{\partial t} - \hat{H}|\Psi(t)\rangle + \int_{t_1}^{t_2} dt\, \langle (i\frac{\partial}{\partial t} - \hat{H})\Psi(t)|\delta\Psi(t)\rangle + i\langle\Psi(t)|\delta\Psi(t)\rangle\,|_{t_1}^{t_2} = 0 \tag{22}$$

Note that here we have adopted slight notational changes (e.g. using the times $t_1, t_2$ rather than $0, T$). Actually, the last term on the r.h.s. vanishes because $\delta\Psi(t)$ vanishes at the boundaries of the time interval. For the sake of a later argument it is retained in Eq. (I-6). Eq. (I-6) implies that $Re\,\langle \delta\Psi(t)|i\frac{\partial}{\partial t} - \hat{H}|\Psi(t)\rangle = 0$ (see Eq. 10), but mistakenly it is stated that " the vanishing of the first two terms [in Eq. I-6] is equivalent to the td SE". Here the possibility is disregarded that $\langle \delta\Psi(t)|i\frac{\partial}{\partial t} - \hat{H}|\Psi(t)\rangle$ may vanish (for all allowed variations within a given parametrization of the wave function) while $(i\frac{\partial}{\partial t} - \hat{H})|\Psi(t)\rangle$ does not.

Next the discussion focusses on the RG AIF and the corresponding VP for the td density (see Eqs. 1 and 13). It is stated that $\delta A[n] = 0$ readily implies Eq. (I-9), reading

$$V(\boldsymbol{r}, t) = \frac{\delta A_0[n]}{\delta n(\boldsymbol{r}, t)} \tag{23}$$

Obviously, this is just a more explicit form of Eq. (17) obtained by expressing $\delta A[n]$ in terms of a functional derivative with respect to the td density. To make contact between the two forms given by Eq. (I-9) and Eq. (17) let us note that the specific (external) potential,

$$V[n] = \int d\boldsymbol{r} \int_{t_1}^{t_2} dt\, V(\boldsymbol{r}, t) n(\boldsymbol{r}, t) \tag{24}$$

can be treated separately in a partitioning of the $A[n]$ into a universal and a specific part, $A[n] = A_0[n] - V[n]$. This yields

$$\frac{\delta A[n]}{\delta n(\boldsymbol{r}, t)} = \frac{\delta A_0[n]}{\delta n(\boldsymbol{r}, t)} - V(\boldsymbol{r}, t) \tag{25}$$

However, as discussed above it has been tacitly assumed here that the integrand in $A[n]$ at a given time $t$ depends only on $n(t)$. If $\delta A[n]$ can be expressed in terms of functional derivatives at all, then one has to expect that functional derivatives with respect to $\dot{n}(t)$, or even $\ddot{n}(t)$ will come into play. So irrespective of the problem of the definiteness of $A[n]$, Eq. (I-9) certainly is not appropriate.

While Eq. (I-9) is seen as questionable, the origin of the problem is located not in the AIF but rather in the VP, implying that the remedy should be a modification of the latter. So let us see what the "solution" here is.

Performing the algebra along Eqs. (I-5,I-6) in the case of $\delta A[n] = 0$, the analogue to Eq. (I-5) reads

$$\delta A[n] = \int_{t_1}^{t_2} dt\, \langle \delta\Psi[n,\delta n](t)|i\frac{\partial}{\partial t} - \hat{H}|\Psi[n](t)\rangle + \int_{t_1}^{t_2} dt\, \langle \Psi[n](t)|i\frac{\partial}{\partial t} - \hat{H}|\delta\Psi[n,\delta n](t)\rangle = 0 \tag{26}$$

where the variations of the td density vanish at the initial and final times, $\delta n(t_1) = \delta n(t_2) = 0$. As mentioned above, Vignale rightly observes that the corresponding variation of the wave function may not vanish at the final time, that is,

$$\delta\Psi[n,\delta n](t_2) \neq 0 \tag{27}$$

Integration by parts then leads to to the result

$$\delta A[n] = 2\int_{t_1}^{t_2} dt\, Re\, \langle \delta\Psi[n,\delta n](t)|i\frac{\partial}{\partial t} - \hat{H}|\Psi[n](t)\rangle + i\langle \Psi[n](t_2)|\delta\Psi[n,\delta n](t_2)\rangle = 0 \tag{28}$$

where in contrast to the VP for the td wave function there is the possibly non-vanishing extra term $\langle \Psi[n](t_2)|\delta\Psi[n,\delta n](t_2)\rangle$ on the r.h.s.. Apparently, consistency with the exact solution of the td SE can only be achieved if the first term on the r.h.s. of Eq. (28) vanishes. This seems to imply that the original VP is not valid and must be replaced by

$$\delta A[n] - i\langle \Psi[n](t_2)|\delta\Psi[n,\delta n](t_2)\rangle = 0 \tag{29}$$

It appears though that this conclusion is somewhat premature.

What Eq. (28) shows is that the AIF VP for the td density is not equivalent to a VP of the (Dirac-Frenkel) form

$$Re\, \langle \delta\Psi[n,\delta n](t)|i\frac{\partial}{\partial t} - \hat{H}|\Psi[n](t)\rangle = 0 \tag{30}$$

(Moreover, one may not assume that $i\delta\Psi[n,\delta n](t)$ is an allowed variation, so that there is no analogue of Eq. (30) for the imaginary part.) While this is an interesting finding, it does not yet discredit the original AIF VP. Let us again recall the idea underlying the AIF VP: to find a density trajectory $n(t)$ such that the associated wave function $\Psi[n](t)$ minimizes (makes stationary) the expectation value of the td SE deviation operator in a given time interval $[t_1, t_2]$, where the manifold of allowed variations of $n(t)$ is restricted by $\delta n(t_1) = \delta n(t_2) = 0$. There is no *a priori* reason why this should not be consistent with an exact solution of the td SE. Let $n(t)$ be the density trajectory associated with the exact solution of the td

SE $\Psi[n](t)$ (for a given initial value $\Psi(t_1)$). Obviously, then the first term on the r.h.s. of Eq. (28) vanishes, but the disturbing 2nd term could vanish here as well, because for this particular $n(t)$ all variations $\delta\Psi[n,\delta n](t_2)$ consistent with $\delta n(t_2) = 0$ might be orthogonal to $\Psi[n](t_2)$, so that

$$\langle\Psi[n](t_2)|\delta\Psi[n,\delta n](t_2)\rangle = 0 \tag{31}$$

Of course, this is not a positive proof for the consistency of the AIF VP with an exact solution of the td SE (a valid proof would presuppose the definiteness of the AIF). However, it discloses a highly plausible loophole that has been entirely disregarded in I. This means that the conclusion concerning the necessity of a modified VP is not stringent.

One might argue that the extra term in Eq. (29) does not do harm because it will (possibly) vanish anyway for the exact td density. However, in the case of a non-exact AIF it would distort the the solution from the optimal one. Moreover, with regard to deriving an EOM the constant extra term is just a nuisance. It forces one to introduce the functional derivative of a ($N$-electron) wave function,

$$\Psi[n](\boldsymbol{r}_1,\ldots,\boldsymbol{r}_N,t_2) = \int d\boldsymbol{r}\int dt\,\frac{\delta\Psi[n](\boldsymbol{r_1},\ldots,\boldsymbol{r}_N,t_2)}{\delta n(\boldsymbol{r},t)}\delta n(\boldsymbol{r},t) \tag{32}$$

The functional derivative in the integrand is an awesome entity indeed, even if needed only for performing the overlap integral in Eq. (I-12), which according I represents the final EOM. Let us note that here, as above, the possible emergence of more general functional derivatives is ignored.

To summarize, there are two major misconceptions in paper I. First, it is supposed (without discussion) that the RG AIF can be expressed entirely in terms of functional derivatives with respect to the density. While this goes back to the original RG paper [1], it is certainly inadequate. One has to expect that functional derivatives with respect to the first (and possibly higher) time derivatives of the td density come into play. So any conclusions based on a too narrow representation in terms of density functional derivatives will be flawed.

Second, the conclusion that the original AIF is not consistent with the exact solution of the td SE is not compelling. There may be a plausible way out of Vignale's dilemma, so that there is no need for a modification of the original VP. The extra term advocated in I not only is incompatible with the intuitive idea of the AIF but leads, moreover, to a hopeless complication in the aspired derivation of an EOM for td denities.

## VI. CONCLUDING REMARKS

The problems associated with the use of the RG AIF have a simple and rather obvious reason, namely, that it is not well-defined in the first place. The indefiniteness with respect to an arbitrary constant due to the undetermined td phase in the wave functions does not disappear in the variational procedure. In fact, Vignale's observation that the density induced variation of the wave function does not vanish at the final time, makes the latter strikingly manifest. One has to face the conclusion that the RG AIF is ill-defined and cannot be used as a means to derive a density-based EOM.

Given that fundamental deficiency, one will not expect anything meaningful to emerge from merely modifying the VP, as proposed in I. Generally speaking, a logically stringent argumentation based on a false premise could be useful in laying bare the defect of the premise. However, as we have shown, the arguments in paper I are themselves hardly rigorous and partly even misleading.

Accepting the fact that the RG AIF is incurably ill-defined, one is referred back to the direct mapping-based foundation of TDDFT, a supposedly valid alternative (see, for example, Marques and Gross [17]). However, one may wonder how the essentially mathematical RG1 mapping theorem could suffice to establish a physical (and causal) EOM for the time-evolution of a quantum system (see Refs. [6, 7]). Unfortunately, there is no second founding paper (replacing the discredited Ref. [1]), in which the mapping-based route to time-dependent Kohn-Sham equations is fully disclosed and, thus, could serve as the basis for a critical analysis. A crucial ingredient of the mapping foundation, namely, the fact that the td Kohn-Sham equations and the exchange correlation potential-functional herewith introduced can only be established as a fixed-point iteration scheme - though one not backed by a valid VP [7] - still has the status of an unpublished insider lore.

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